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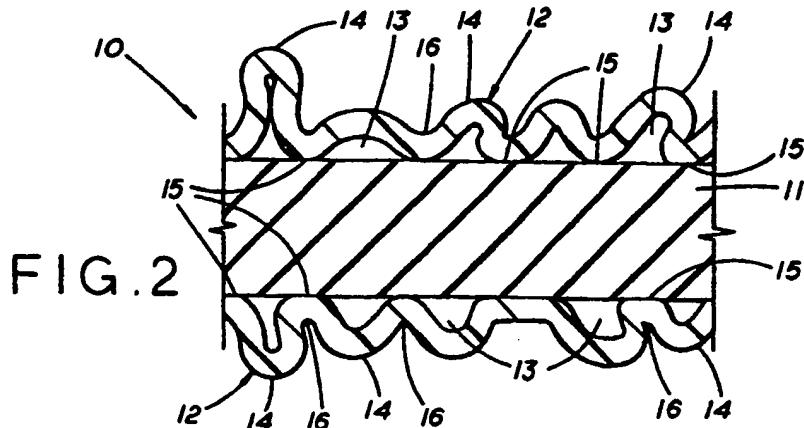
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(54) Low gloss film and process of manufacture.

(57) A multilayer film comprises a elastomeric core layer (11) and at least one thermoplastic skin layer (12) having a microundulating surface configuration providing the film with a low gloss.



LOW GLOSS FILM AND PROCESS OF MANUFACTURE

BACKGROUND OF THE INVENTION

This invention relates generally to films useful in a variety of domestic applications in which the properties of elasticity, low gloss, and good "hand" are desirable. Examples of such applications include back sheets for diapers, colostomy bags, elastic garment closures (e.g. sleeve bands, diaper leg bands and waist band), and the like. In one aspect the invention relates to a coextruded film and method of manufacture.

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DESCRIPTION OF THE PRIOR ART

Although the film of the present invention, because of its unique properties, may be used in a variety of applications, the disclosure herein will emphasize its application as a diaper back sheet. Film used in diapers and similar absorbent goods such as incontinent garments must possess certain physical properties and certain esthetic properties for successful marketing. For example, the backsheet of diapers which contain the absorbent material must possess adequate physical properties for durability. Equally important is the appearance of the diaper for customer appeal. Experience has shown that the diaper must not only possess a matte finish (low gloss) but must also possess the proper "hand" or "feel", and be free of "plastic" noise on handling. The term "hand" and "feel" are frequently used in the textile industry to describe a property of fabric which affects the sense of touch (e.g. rough, plastic, silky, coarse, etc.). Moreover, it is important that the diaper fit comfortably on the wearer and provide secure containment.

The conventional procedure for obtaining matte finish diaper back sheets is by a process in which a cast film is passed through the nip of a pair of rollers, one of which has a pattern engraved roller or a special sanded finish. The film is thus embossed with either a pattern finish or irregular sanded finish, both of which provide low gloss on the film processed thereby. The conventional embossing process described above has limitations, particularly with regards to economics. While modern cast lines can produce films at rates up to 1500 feet per minute, the embossing process is limited to line speeds of about 700 feet per minute. Moreover, the "hand" or "feel" of the embossed film is not particularly good.

Low gloss or matte finish films are disclosed extensively in patents and the published literature. See for example Japanese Kokai's 76 31,773, 76 31,1774, 81 11,231, GB 1453649, US 3,843,478, and US 4,522,887.

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SUMMARY OF THE INVENTION

It has been surprisingly discovered that a low gloss, elastic, matte finish film possessing exceptional hand and feel is obtained by a coextruded multilayer film comprising an elastomeric core (layer B) and thermoplastic skins (layers A). The core layer B has a substantially flat disposition whereas the thermoplastic skin layers A have a microundulating or rippling configuration. The microundulating configuration not only imparts a matte, low gloss finish to the surface of the composite, but also improves the film's tactile properties (i.e. "hand" and "feel"). Moreover, the rubber core layer of the composite imparts elasticity to the garment thereby providing exceptional fit and containment (if used in a diaper).

The process for producing the film includes generally the steps of (a) coextruding an ABA composite film wherein the skin layers A comprise thermoplastic material and the core layer B comprises elastomeric material; (b) stretching the composite film at least 100% of its original length; and (c) relaxing the stretched film. This process produces a film with the skin layers having microundulations described above. Relaxation of the stretched film causes the rubber core layer to contract more than the thermoplastic skin layers thereby providing an irregular microsurface on the composite structure. In the relaxed stretched condition, the microundulations, while not readily discernable by the naked eye, impart a low gloss and silky feel to the film. (The term "relaxed stretched condition", as used herein, describes the condition of the film after the stretching and relaxation steps of the process.)

The elastomeric core for the ABA structure described above may comprise and/or include a variety of

elastomers. The preferred elastomers are polyisobutylene (PIB), butyl rubber, ethylene-propylene copolymer rubber (e.g. EPM or EPDM), block copolymer rubbers (e.g. SBS, SIS, or SEBS), and mixtures of these copolymers. The skin layers are thermoplastics, preferably ethylene copolymers such as ethylene vinyl acetate (EVA) copolymers.

5 The relative thickness of the A and B layers of the ABA composite may range within wide limits so long as the stretch/ contraction step produces the microundulations which provide a gloss of less than 20, preferably 10 or less. The preferred ranges are as follows:

10	<u>BROAD RANGE</u>		<u>PREFERRED RANGE</u>
	<u>wt% of Total Thickness</u>		<u>wt% of Total Thickness</u>
15	A	2.5 to 20	5 to 15
	B	95 to 60	90 to 70
	A	2.5 to 20	5 to 15

20 The total thickness of the multilayer film of the present invention is less than 20 mils and preferably 10 mils or less.

25 The microundulation configuration of the skin layers of the film according to the present invention are to be distinguished from the embossing or sanded finish of the prior art. The irregular surface provided by the former is a result of gathering on a micro scale whereas the latter achieves surface pattern or irregularities by permanent deformation of the film.

BRIEF DESCRIPTION OF THE DRAWINGS

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Figure 1 is a cross sectional view of the three layer film of the present invention.

Figure 2 is a cross sectional view of a magnified portion of the film shown in Figure 1.

Figure 3 is a magnified top plan view of a portion of the film shown in Figure 2, illustrating the irregular surface.

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DESCRIPTION OF THE PREFERRED EMBODIMENT

With reference to Figure 1, the film 10 of the present invention is a composite comprising an 40 elastomeric core layer 11 and two thermoplastic skin layers 12. This ABA film composite may be manufactured by coextrusion processes (as described in more detail below) and by selective stretching and relaxing to produce the desired surface profile of layers 12 (illustrated in Figure 2). The film 10 thus produced surprisingly exhibits the following properties which make it ideal for a number of applications:

- (1) good strength properties (in both TD and MD)
- (2) matte finish
- (3) good hand and feel (silky)
- (4) quiet nonplastic "noise" (i.e. rustle free)
- (5) high elasticity

As described in more detail below, the compositions of the elastomeric core layer 11 and thermoplastic 50 skin layers 12 may vary within relatively wide limits, provided that the core and skin layers exhibit differential contraction to produce the microundulating surface. The terms "rubber" and "elastomers" are used interchangeably herein and fit the definition of rubber per ASTM D 1566 which is incorporated herein by reference. Also, the "MD" and "TD" are abbreviations for machine direction and transverse direction which are used in their conventional sense meaning, respectively, the direction in which the film is 55 processed and the direction at right angles therewith.

Elastomeric Core Layer:

The rubber core layer 11 must be composed of a rubber which (a) is capable of melt extrusion in the same general temperature range of thermoplastic resin used in the skin layers; (b) can be extended at least 100% of its original length and recover a substantial amount of its extended length (preferably more than 50%); (c) adhere to the skin layer; and (d) permit random separation from the skin at locations generally uniformly distributed across its surface to produce the microundulating surface profile.

The rubbers (elastomeric resins) which can be used in the practice of this invention include both synthetic and natural rubbers; preferably the rubbers have a glass transition temperature (Tg) of less than 0 °C and have a Shore A hardness of 50 or below, preferably 40 or below, or a 100% modulus of 110 kg./cm² or less or a Young's modulus below 1000 kg./cm². Illustrative, non-limiting examples of rubbers suitable for use in the practice of this invention include polyisobutylene (PIB), butyl rubber, halogenated butyl rubber, ethylene propylene rubber (EPM), ethylene-propylene diene rubber (EPDM), polyisoprene, polychloroprene, styrene-butadiene rubber, polybutene copolymers, nitrile rubbers, chlorosulfonated polyethylene, block copolymer rubber, etc.)

The block copolymer rubber includes from 50 to 85 wt % of a central block of a rubbery olefin polymer of generally equal proportions of ethylene and butylene units and terminal blocks of polystyrene (SEBS). A commercially available block copolymer rubber is Kraton sold by Shell Oil Company. Other rubbery copolymers utilize a central block of butadiene (SBS) or isoprene (SIS) instead of the ethylene butylene copolymers. The preferred block copolymer rubbers are the SEBS copolymers having a molecular weight of between about 50,000 to 120,000.

The terms EPM and EPDM are used in the sense of their ASTM designations (ASTM D-1418-72a). EPM is an ethylene-propylene copolymer which can be cross-linked by radiation curing or peroxide curing.

As used in the specification and claims the term "EPDM" means terpolymers of ethylene alphaolefin and non-conjugated diene. The non-conjugated diolefin can be straight chain, branched chain or cyclic hydrocarbon di-olefins having about 6 to about 15 carbon atoms such as:

- 25 A. straight chain dienes such as 1,4-hexadiene and 1,6-octadiene;
- B. branched chain acrylic dienes such as 5-methyl-1, 4-hexadiene; 3,7-dimethyl-1,6-octadiene; 3-7-dimethyl-1, 7-octadiene and the mixed isomers of dihydro-myricene and dihydroocinene;
- C. single ring alicyclic dienes such as 1,3-cyclo-pentadiene; 1,4-cyclohexadiene; 1,5-cyclo-octadiene and 1,5-cyclododecadiene;
- 30 D. multi-ring alicyclic fused and bridged ring dienes such as tetrahydroindene, methyl, dicyclopentadiene; bicyclo-(2,2,1)-hepta-2, 5-diene; alkenyl, alkylidene, cycloalkenyl and cycloalkylidene norbornenes such as 5-methylene-2-norbornene (MNB), 5-propenyl-2-norbornene, 5-(4-cyclopentenyl)-2-norbornene, 5-cyclo-hexylidene-2-norbornene, 5-vinyl-norbornene and norbornadiene.

Of the non-conjugated dienes typically used to prepare EPDM terpolymers the preferred dienes are dicyclopentadiene, 1-,4-hexadiene, 5-methyl-2-norbornene and 5-ethylidene-2-norbornene. Particularly preferred diolefins are 5-ethylidene-2-norbornene (ENB) and 1,4-hexadiene.

EPDM elastomers and their general method of manufacture are well known in the art. The preferred EPDM elastomers contain about 20 to about 90 wt% ethylene, more preferably about 30 to 80 wt% ethylene, most preferably about 35 to about 75 wt% ethylene.

40 The alpha-olefins suitable for use in the preparation of EPDM are preferably C₃-C₁₆ alpha olefins. Illustrative non-limiting examples of such alpha olefins are propylene, 1-butene, 1-pentene, 1-hexene, 1-octene and 1-dodecene. The alpha olefin is generally incorporated into the EPDM polymer at about 10 to about 80 wt%, more preferably at about 20 to about 70 wt%. The non-conjugated diene is incorporated into the EPDM at about 0.5 to about 15 wt%; more preferably about 1 to about 5 wt%, e.g., 3 wt%.

45 The term "nitrile rubber" means an acrylonitrile copolymer rubber. Suitable nitrile rubbers comprise rubbery polymers of 1,3-butadiene and about 20-50 wt% acrylonitrile. Any nitrile rubber which is a "solid" rubber having an average molecular weight of at least 50,000 and preferably between about 100,000-1,000,000 can be used. Commercially available nitrile rubbers suitable for the practice of the invention are described in Rubber World Blue Book, 1980 Edition, "Materials and Compounding Ingredients for Rubber", 50 pages 386-406, which is incorporated herein by reference.

Butyl rubber is a copolymer of an isoolefin and a conjugated multiolefin. The useful copolymers comprise a major portion of isoolefin and a minor amount, preferably not more than 30 wt%, of a conjugated multiolefin. The preferred copolymers comprise about 85-99.5 wt% (preferably 95-99.5 wt%) of a C₄-C₇ isoolefin, such as isobutylene, and about 15-0.5 wt % (preferably about 5-0.5 wt%) of a multiolefin of about 4-14 carbon atoms. These copolymers are referred to in the patents and literature as "butyl rubber"; see, for example, the textbook Synthetic Rubber by G.S. Whitby (1954 edition by John Wiley and Sons, Inc.), pages 608-609, etc. which is incorporated herein by reference. The term "butyl rubber" as used in the specification and claims includes the aforementioned copolymers of an isoolefin having 4-7 carbon

atoms and about 0.5 to 20 wt% of a conjugated multiolefin of about 4-10 carbon atoms. Preferably these copolymers contain about 0.5 to about 5% conjugated multiolefin. The preferred isoolefin is isobutylene. Suitable conjugated multiolefins include isoprene, butadiene, dimethyl butadiene, piperylene, etc.

Commercial butyl rubber is a copolymer of isobutylene and minor amounts of isoprene. It is generally prepared in a slurry process using methyl chloride as a vehicle and a Friedel-Crafts catalyst as the polymerization initiator. The methyl chloride offers the advantage that AlCl_3 , a relatively inexpensive Friedel-Crafts catalyst is soluble in it, as are the isobutylene and isoprene comonomers. Additionally, the butyl rubber polymer is insoluble in the methyl chloride and precipitates out of solution as fine particles. The polymerization is generally carried out at temperatures of about -90C to -100C. See U.S. Patent Nos. 2,356,128 and 2,356,129 incorporated herein by reference.

In the halogenation process, butyl rubber in solution is contacted with chlorine or bromine in a series of high-intensity mixing stages. Hydrochloric or hydrobromic acid is generated during the halogenation step and must be neutralized. For a detailed description of the halogenation process see U.S. Patent Nos. 3,029,191 and 2,940,960, as well as U.S. Patent No. 3,099,644 which describes a continuous chlorination process, all of which patents are incorporated herein by reference.

The rubbers used in the practice of this invention are preferably utilized in their unvulcanized state.

The preferred elastomeric core of the film comprises a blend of ethylene propylene copolymer rubber or EP diene rubber (EPM or EPDM) and butyl type rubber (PIB or butyl rubber) in weight ratios ranging from 3:7 to 7:3, preferably 6:4 to 4:6. Although EP or butyl satisfy the extension properties, the preferred elastomeric core is comprised of a blend to provide a balance of properties. The crystallinity of the EP rubber confers pellet stability to the blend (i.e. resists agglomeration during processing) and the butyl type rubber imparts low modulus (i.e. softness) to the film.

The core may include other additions such as slip agent, block copolymer rubber (e.g. Kraton), antioxidant, or plasticisers (e.g. refined oil).

The rubber compound ingredients may be blended in the proper weight ratio prior to introduction into the extruder.

Thermoplastic Skin Layers:

Thermoplastics are distinguished from elastomers by their substantially inelastic properties. Whereas certain thermoplastics such as EVA exhibit some elasticity, the degree of elasticity falls short of that required to meet the "rubber" definition of ASTM D 1566. In general, any of the thermoplastic resins known in the prior art to be useful as films may be used as the skin layers, provided the film extrudate bonds satisfactorily to the rubber core layer. Suitable thermoplastic polymeric resins include polymers of branched and straight chained olefins such as polyethylene, polypropylene, polybutylene, polypentene, polymethylpentene and the like and various copolymers of ethylene and copolymers of propylene.

The ethylene copolymers are particularly preferred for the skin because they adhere reasonably well to rubber and possess some elasticity, and do not detract from the overall elasticity properties of the ABA film.

The ethylene copolymers include those of ethylene and at least 5 wt % (preferably at least 10 wt %) of unsaturated esters of lower carboxylic acids or unsaturated carboxylic acids, and an alpha-olefin having 3 to 12 carbon atoms, preferably 3 to 8 carbon atoms. The preferred ethylene comonomers include butene, hexene, and vinyl acetate. Specific preferred thermoplastics are ethylene vinyl acetate (EVA) copolymers, ethylene acrylic acid (EAA) copolymers, ethylene butyl acrylate (EBA) copolymers, ethylene methacrylate (EMA) copolymers, and low density ethylene butene or hexene copolymers (comonomers wt % of 5 to 40 and preferably of 10 to 35 wt %). The ethylene copolymers generally contain from 60 to 95 weight percent ethylene, most preferably from 65 to 90 weight percent ethylene.

The most preferred ethylene copolymer useful in the present invention is EVA. EVA may have a vinyl acetate (VA) content between about 5% and 40% by weight, with about 10 to 35% by weight being preferred and 15 to 25% by weight being most preferred.

VA contents below about 5 wt % do not confer sufficient flexibility and orientability for purposes of the present invention and VA contents above 40 wt% exhibit excessive tackiness. The best balance of orientability and non-tackiness occurs at VA contents between 15 and 25 wt %.

Preferred Melt Index (ASTM-D-1238, Condition E) for EVA is from 0.5 to 20, with 1 to 10 being preferred and 1-5 most preferred.

It will be appreciated that the composite film may have an AB or ABC multilayer structure as well as an ABA structure described above. In the ABC multilayer, the skin layers A and C may have a different composition with skin layer A having the composition as described above. In this embodiment, the film can

be formulated so that only layer A will have the desire surface profile described above.

Film Manufacture:

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The ABA film of the present invention may be manufactured by a three step process:

- (1) The elastomeric resin and thermoplastic resins are coextruded to form the ABA (or ABC) composite structure. (Since no embossing rollers are required, higher line speeds are possible.)
- (2) The composite film is stretched preferably in the machine direction (this stretches the ABA layers equally).
- (3) The film is relaxed causing the core layer to contract much more than the skin layers, producing a rippled surface on each skin layer.

Following Step 1, the film generally has three layers bonded together and exhibits a relatively high degree of transparency since the outer surfaces of the skin are relatively smooth. The stretching of Step 2 may even increase the transparency since it will tend to reduce this film gauge and smooth the film surface further.

The contraction of Step 3, surprisingly, causes the film to become hazy and imparts a silky feel to the surface. As shown in Figure 2 (which is a drawing of a microphotograph of the film magnified 5000 times), the contraction of the elastomeric core 11 causes the thermoplastic skins 12 to separate from the surface of core 11 at spaced locations, the separations being shown as 13. This produces a rippling surface on the skins 12 wherein the ripples 14 extend generally in the transverse direction (as shown in Figure 3) with respect to the stretch direction. It is this rippling, irregular surface on a micro scale which provides the matte finish and smooth silky feel. The irregular surface scatters light and the reduced area contacted by the hand gives the film a silk-like feel.

Upon stretching the film again the undulations or ripples tend to disappear permitting light to be reflected uniformly from the film surface, restoring transparency.

The coextrusion of the ABA composite may be performed using conventional blending, compounding, and extruding equipment.

The stretching step may be carried out by a number of techniques. For example, with cast film two Godet rolls operating at different speeds may be used to stretch the film in the machine direction. A tenter frame may be used to stretch the film in the transverse direction. The film may also be bi-directionally stretched in which case the surface undulations may intersect.

With blown film, the stretching may be achieved by the "double bubble" process which involves producing a normal blown stalk, collapsing the bubble, and reinflating the bubble to a larger diameter.

Stretching may be carried out one or more times. From one to three stretchings will be satisfactory for most films.

The degree of stretching will depend largely on the composition of the film but will be between 100% to 700% of original length, with 200 to 500% being preferred and 300% to 400% most preferred. The stretching preferably is carried out at room temperatures (65 to 80 °F).

The relaxation step may be carried out as follows. With Godet rollers, nontension take up rollers may be employed. With the tenter frame, the tenter track may be positioned to return the film to the nontension state following stretching. With blown film, relaxation is achieved by merely collapsing the bubble. The film will reach its fully relaxed state in about 5 to 10 minutes following release of the tension.

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Film Properties:

The film may be characterized by dimensional profile, gloss, coefficient of friction and secant modulus.

The dimensional profile of the surface of skin layer 12 may be measured by a number of instruments capable of precise surface roughness measurements. One such instrument is Surtronic 3 manufactured by Rank Taylor Hobson. This instrument is capable of measuring in the range of 0 - 1000 micro inches at accuracy of $\pm 2\%$ of the reading.

Gloss is a measure of film transparency and is determined by ASTM D 1003-67. Gloss is represented by a numerical value range of 0-100. Films with gloss readings below 20 are low to moderate gloss films and those with readings below 10 are preferred low gloss films. Matte finish for applications such as diaper back sheets will have gloss readings of 10 or less.

Coefficient of friction (COF) were determined by ASTM D1894-78.

The tensile and tear strengths and secant modulus (which indicates softness) were determined by

standard ASTM tests.

EXPERIMENTS

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Materials:

10 Samples of ABA film were prepared by coextruding core resin compound with skin resin using a Killion extruder. The core layer and skin layers had the compositions shown in Table I (all weights are wt %).

TABLE I

15	FILM SAMPLE	CORE LAYER	EACH SKIN	ABA THICKNESS (%)	AVG. FILM THICKNESS (MILS)
20	A	EPM ¹ (50%) Butyl Rubber ⁴ (50%) MI = 0.19	EVA ⁶	9/29/9	3.4
25	B	EPM ² (50%) Butyl Rubber ⁴ (50%) MI = 0.20	EVA ⁶ EVA ⁶	9/82/9	3.3
30	C	EPM ³ (40%) Butyl Rubber ⁴ (40%) Block Copolymer ⁵ (20%) MI = 0.23	EVA ⁶	9/82/9	2.4
35	D	EPM ³ (35%) Butyl Rubber ⁴ (35%) Block Copolymer ⁵ (30%) MI = 0.21	EVA ⁶	9/82/9	3.2
	E	EPM ² (50%) Butyl Rubber ⁴ (50%) MI = 0.2 - 0.3	EVA ⁷ (90%) PE ⁸ (10%)	10/80/10	4.8

1 - Vistalon 3708 sold by Exxon Chemical Company

2 - Vistalon 7000 sold by Exxon Chemical Company

3 - Vistalon 719 sold by Exxon Chemical Company

4 - Butyl 065 sold by Exxon Chemical Company

5 - Kraton G-1652 sold by Shell Chemical Company

6 - LD 702.45 (0.3 MI) sold by Exxon Chemical Company 12% VA with small amounts of BHT, antiblock and slip additive.

7 - LD 721.62 (2.5 MI) 18% VA with small amounts of slip agent and antiblock.

8 - PE containing minor amounts of slip agent and antiblock additive.

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The core layers of samples A,B,C, D and E also included small amounts of slip and antiblock additives. The MI of each core layer compound was determined at condition E and the units are dg/min at 190 °C.

50 The extruder conditions were as shown in Table II.

TABLE II

	<u>Skin Extruder</u>	<u>Core Extruder</u>
5	Size (inches)	0.75
	L/D	24/1
	HP	1.5
	Comp. Ratio	3/1
10	Extruder Temp. (° F)	350° - 430°
	Melt (° F)	365° - 375°
	RPM	30
	GMS/min	14
		1
		24/1
		3.0
		3/1
		350° - 430°
		419° - 422°
		80
		62.5 - 65.5

15 The film samples had the following physical properties (before stretching):

TABLE III

	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>
20	Tensile Strength, psi (MD)	1135	1200	1400
	Ultimate Elongation, % (MD)	410	400	440
	1% Secant Modulus, Psi (MD)	2500	2500	4200
25	Tear Strength, g/mil (MD)	41	34	70
				90

Each sample having length of 0.591 inches was tested on a Instron with maximum jaw separation of 2.5 inches and operated at a speed of 5 in./min.

30 The test procedure was as follows:

- Stretch sample to 100% or 150% elongation and immediately remove stress.
- Stretch sample three times.

Data (presented in Table IV)

- Modulus, psi

35 - Permanent Set after 3rd Cycle; % (unrecovered strain at return to zero stress divided by maximum strain after 5 min. recovery period).

(Permanent set indicates the amount of recovery from maximum extension.)

- Gloss, before and after Stretching

(All data taken with film sample in relaxed condition.)

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TABLE IV

	FILM SAMPLE			
	<u>A</u>	<u>B</u>	<u>C</u>	<u>D</u>
45	100% Extension -			
	Modulus, psi	345	390	575
	Permanent Set, % (1)	17	18	10
50	60° Gloss			
	Unstretched	34	39	56
	Relaxed Stretched Cond.(1)	5	6	45
			5	8

55 (1) After third cycle

stretching) of these films are presented in Table V.

TABLE V

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		SAMPLE E	
		4-mil	8-mil
<u>Tensile Strength , psi</u>			
	@ Yield MD	225	210
	TD	155	160
	@ Break MD	1,205	960
	TD	705	650
<u>Elongation , %</u>			
	@ Yield MD	30	35
	TD	17	20
	@ Break MD	485	545
	TD	645	600
	<u>1% Secant Modulus , psi MD</u>	1,765	1,355
	TD	1,715	1,480
<u>Elmendorf Tear Strength, g/mil</u>			
	MD	NT ⁽¹⁾	NT
	TD	162	286
	Haze, % - As Made	7.7	17.8
	- After RT Stretching	95.0	95.5
<u>Permanent Set , %</u>			
	3 cycles to 100% Extension - MD	16	12
	TD	16	16
	3 cycles to 150% Extension - MD	28	28
	TD	48	44

⁽¹⁾ NT = No Tear

Sample E films were also tested to determine the effect of stretching on the film secant modulus and COF. The stretched samples were cycled about 3 or 4 times at 200 - 400% of original length. The data are presented in Table VI.

TABLE VI

	4 mils		8 mils	
	Unstretched	Relaxed Condition	Unstretched	Relaxed Condition
5	1% Secant Modulus, psi			
10	MD	1765	720	1355
	TD	1715	370	1480
15	COF			
	MD	>1	0.10	0.58
	TD	>1	0.19	>1
	Gloss	47.2	4.0	59.9
				0.16
				0.58
				10.4

20 The surface profiles of Sample E film were measured using the Surtronic 3 instrument. Table VII presents the surface profile data. (Note stretched film data was after three cycles of 100% elongation and relaxation.)

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TABLE VII

	4-mil Film				8-mil				
	Relaxed		Stretched		Relaxed		Stretched		
	Unstr.		Cond.		Unstr.		Cond.		
	MD	TD	MD	TD	MD	TD	MD	TD	
5									
10									
	Ra, microinches	50	24	96	84	37	25	70	73
15	Rt, microinches	273	163	654	334	189	149	286	621
	Rtm, microinches	182	94	481	274	145	64	223	367
20	Rpm, microinches	92	48	247	145	64	34	115	191
	Pc peak/cm @ 75								
	microinches	23	10	62	57	15	15	39	77
25	tp %								
	25	3	3	2	2	1	9	5	1
	50	18	12	0	7	20	25	11	1
25	75	29	47	1	15	38	38	24	1
	100	39	75	-	-	78	78	-	2
	125	-	95	16	21	100	85	57	-
30	150	56	100	-	-	-	98	-	-
	175	-	-	-	-	-	100	80	13
	200	79	-	22	71	-	-	94	-
	250	99	-	-	-	-	-	100	45
35	325	-	-	49	100	-	-	-	80
	400	-	-	66	-	-	-	-	90
	500	-	-	94	-	-	-	-	-100

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5 Ra - Height of roughness irregularities as measured by the
 average value of the departures from a center line drawn
 such that the sum of the areas above the line equals the
 sum of those below the line.

10 Rt - Maximum peak to valley height over the sample assessment
 length.

Rtm - Mean of the maximum peak to valley heights recorded on
 several passes.

15 Rpm - Mean of the maximum profile height above the mean line on
 several passes.

20 Pc - The peak count is the number of local peaks which project
 through a selectable band centered about the mean line. The
 count is determined only over the assessment length though
 the results are given in peaks per cm (or per in).

25 $P_c = \frac{\text{No. of counts}}{\text{Assessment length (cm)}} = \text{Peaks/cm}$

30 tp% - Bearing ratio tp% is the length of bearing surface
 (expressed as a percentage of the assessment length L) at a
 depth p below the high peak.

35 By making a number of measurements at different depths p,
 and plotting p against tp% the bearing ratio (or Abbott
 Firestone curve) can be drawn.

40 The combination of the Ra and Pc or Rpm and Pc values probably are the best measurements for the
 surface profile of the film. Comparing these values for the unstretched film with those of the relaxed
 stretched film clearly shows that both the amplitude (avg.) and the number of peaks increased as a result of
 the stretching and relaxation process.

45 The combination of Pc and tp% data indicate that although the relaxed stretched film has far more
 peaks (above 75) microinches, the bearing ratio is far less than the unstretched film.

It is preferred that the film of the present invention have the following roughness values, at least in one
 direction (MD or TD) and preferably in both directions.

45

	Broad Range	Preferred Range
Pc @ 75 microinches	30 - 100	50 - 100
tp % @ 75 microinches	less than 25% (1-25%)	less than 10% (1-10%)
tp % @ 250 microinches	less than 90% (20-90%)	less than 50% (20-50%)
Ra microinches	50 - 150	60 - 100
Rpm microinches	100 +	100 - 300

55 From the data presented in Tables IV and VI, the following conclusions regarding the effect of
 stretching and relaxing the ABA film having a rubber core and thermoplastics skins may be reached:

- (1) The surface roughness is increased,
- (2) The gloss is decreased,

- (3) The COF is reduced,
- (4) The secant modulus is reduced, and
- (5) The feel of the film is transformed from plastic to silky (observed).

5

Claims

1. A multilayer coextruded film which comprises
 - (a) a core layer of an elastomeric material; and
 - (b) at least one skin layer of a thermoplastic material, said skin layer being secured to said core layer at spaced apart contact areas and having a microundulating surface in the machine direction of transverse direction such that the film has a gloss of less than 20 and determined by ASTM D 1003-67.
2. Film according to claim 1 wherein the microundulations of the skin layer provide a gloss of 10 or less.
3. Film according to claim 1 or claim 2, wherein the microundulations provide the skin layer with a profile having an Rpm in excess of 100 microinches and a Pc of at least 30 peaks/cm in the machine direction or transverse direction wherein Rpm and Pc are in the same direction and wherein

Rpm is the mean of the maximum height profile above the mean line in the machine direction or transverse direction above and below its center line; and

Pc is the number of local peaks which project through a 75 microinch band centered about the mean line of the surface profile of the skin layer in the machine direction or transverse direction.
4. Film according to claim 3 wherein the film profile has a tp% of less than 25% at a p of 75 microinches and a tp% of less than 90% at a p of 300 microinches, wherein tp% is the bearing ratio and is the length of bearing surface expressed as a % of assessment length in the direction of Rpm and Pc at a depth of p below the highest peak and/or wherein the skin layer has an Rpm of at least 150 microinches and a Pc of at least 50.
5. Film according to any of the preceding claims wherein the elastomeric material is selected from the group consisting of PIB, EPM, EPDM, Butyl Rubber, block copolymer rubber, and mixtures of these and the thermoplastic material is a copolymer of ethylene and a comonomer selected from the group consisting of olefins preferably having from 3 to 8 carbon atoms or vinyl acetate, methylacrylate, methacrylic acid, acrylic acid, butyl acrylate, and mixtures of these.
6. Film according to any of the preceding claims wherein two thermoplastic skin layers are secured to opposite sides of the core layer, said skin layers being secured to the core layer at random spaced apart contact areas to provide each skin layer with microundulating configuration in the machine direction or transverse direction, said film having a gloss value less than 20 as determined by ASTM D 1003-67.
7. Film according to any of the preceding claims wherein the or each skin layer is composed of a polyolefin selected from ethylene and propylene homopolymers and copolymers, preferably a copolymer of ethylene and a comonomer selected from the group consisting of vinyl acetate, methacrylic acid, acrylic acid, butyl acrylate, and olefins having from 3 to 8 carbon atoms, said comonomer comprising from 5 to 40 wt % of the copolymer.
8. Film according to claim 7 wherein the olefin is selected from the group consisting of butene and hexene wherein said olefin comprises from 10 to 35 wt % of the copolymer.
9. Film according to any of the preceding claims wherein the thickness of the film is 10 mils or less and/or the core layer comprises from 60 to 95 wt% of the film thickness preferably from 70 to 90 wt% and/or the or each skin layer from 5 to 15 wt%.
10. Film according to any of the preceding claims wherein the film upon being stretched at least 100% of its length has substantially higher gloss than the unstretched condition.
11. Film according to any of claims 1 to 7, wherein the thermoplastic skin layers comprise EVA wherein the VA comprises from 5 to 40 wt % of the copolymer and the core layer comprises a blend of 30 to 70 wt % of EPM and 70 to 30 wt% of butyl rubber.
12. Film according to any of claims 1 to 7, wherein the core layer comprises a rubber selected from the group consisting of EPM, butyl rubber, block copolymer rubber, and blends thereof.
13. Film according to claim 12 wherein the core layer comprises from
 - from 25 to 45 wt % of EPM,
 - from 25 to 45 wt % of butyl rubber, and
14. Film according to claim 12 or 13, wherein the block copolymer rubber is selected from the group consisting of styrene-ethylene-butylene-styrene and styrene-butadine-styrene block copolymer.
15. A process for producing a low gloss film comprising

(a) stretching a multilayer coextruded film comprising a rubber core layer and thermoplastic skin layers at least 100% of its original length and having a gloss in excess of 30 as determined by ASTM D 1003-67; and

5 (b) relaxing the stretched film wherein the core layer contracts more than the skin layers thereby providing the skin layers with a microundulating surface, said film in the relaxed condition having a gloss of less than 20 as determined by ASTM D 1003-67.

16. Process according to claim 15 wherein steps (a) and (b) are repeated at least two times.

17. Process according to claims 15 or 16, wherein step (a) stretches the film from 300 to 500 % of its original length.

10 18. Process according to any of claims 15 to 17, wherein the stretching and relaxation steps increase the film roughness as measured in the stretch direction as follows:

Ra by at least 50%,
Rpm by at least 50%, and
Pc by at least 50%

15 wherein
Ra is the height of roughness irregularities as measured by the average of the departures from a center line drawn such that the sum of the areas above the line equals the sum of those below the line;
Rpm is the mean of the maximum height profile above the mean line; and
Pc is the number of local peaks which project through a 75 microinch band centered about the mean line of
20 the surface profile of the skin layer.

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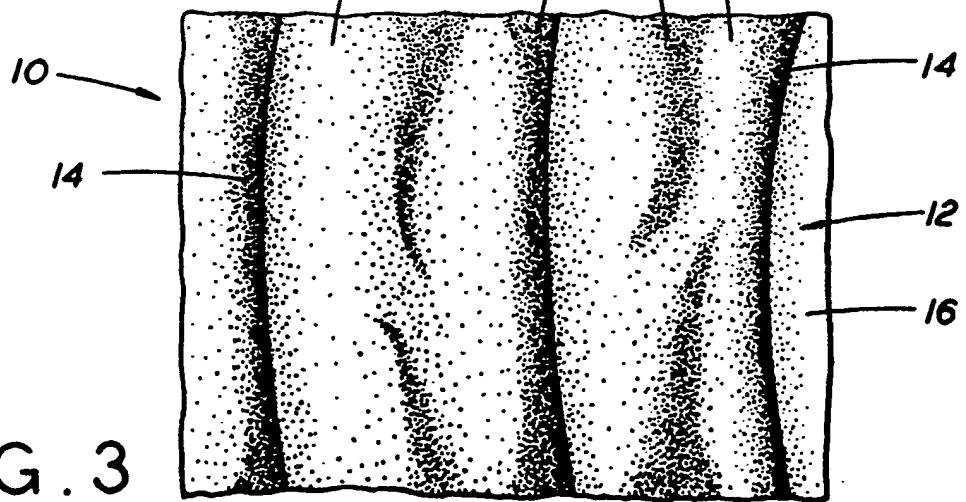
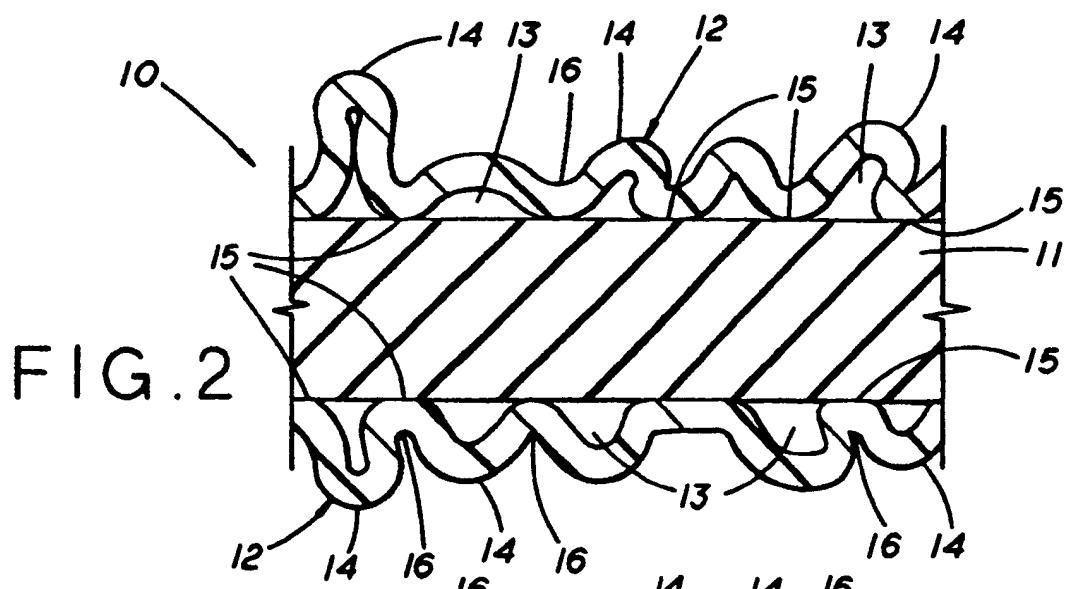
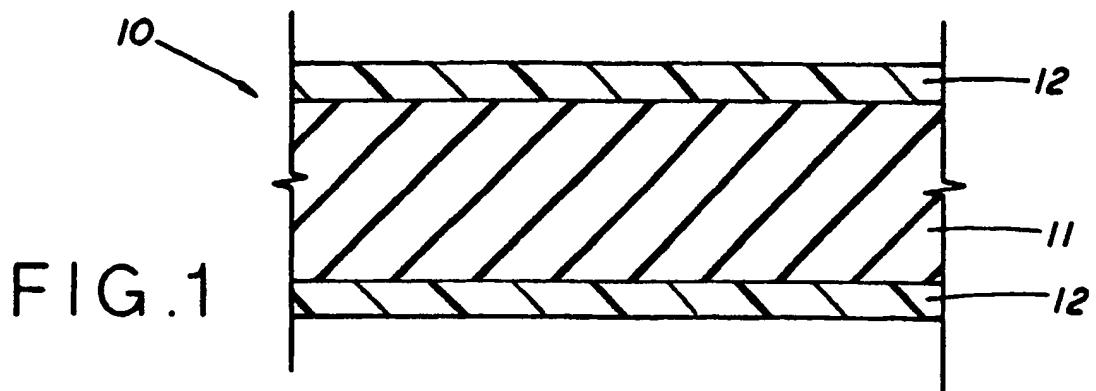
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EP 89 31 0019

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
E	CHEMICAL PATENTS INDEX, DOCUMENTATION ABSTRACTS JOURNAL, Derwent Publications, Ltd, section A plastics, polymers, week 9004, accession no. 90-029991/04, 21st March 1990; & US-A-171 730 (EXXON CHEMICAL PATE) 14-11-1989 ---	1-18	B 32 B 25/08 B 32 B 7/04 B 32 B 3/26 B 32 B 31/16
X	EP-A-0 102 245 (CHICOPEE) * Page 1, lines 7-14; page 2, line 17 - page 3, line 11; page 5, line 3 - page 6, line 5; page 9, lines 1-15; figures 1,2,3,14 *	1,6,7,15	
A	---	5,11	
A	US-A-4 781 966 (TAYLOR) * Column 1, lines 14-26; column 2, lines 31-63; column 6, lines 30-37; column 9, line 64 - column 10, line 42; column 10, line 61 - column 11, line 2; column 11, lines 13-19; figures 1,2 *	1,6,15	
A	US-A-4 606 964 (WIDEMAN) * Column 4, lines 38-47; column 5, lines 15-36; column 6, lines 14-18,30-34; column 7, line 56 - column 8, line 16; figures 3,4; claims 11,12 * -----	1,6,7,15	TECHNICAL FIELDS SEARCHED (Int. Cl.5) B 32 B A 61 F 13
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	08-06-1990	IBARROLA TORRES O.M.	
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